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Climate and productivity affect total mercury concentration and bioaccumulation rate of fish along a spatial gradient of subarctic lakes

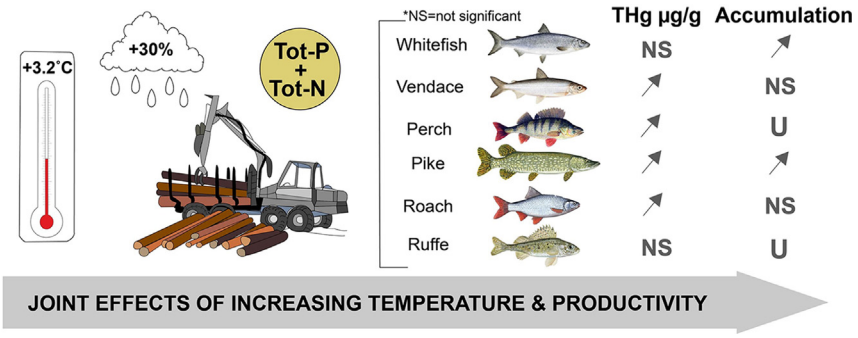
Salla A. Ahonen [a,*], Brian Hayden [b], Jaakko J. Leppänen [a], Kimmo K. Kahilainen [c]

[a] Department of Environmental Sciences, University of Helsinki, P.O. Box 65, FI-00014, Finland
[b] Canadian Rivers Institute, Biology Department, University of New Brunswick, Canada
[c] Faculty of Biosciences, Fisheries and Economics, The Norwegian College Fishery Science, UiT The Arctic University of Norway, 9037 Tromsø, Norway

HIGHLIGHTS

- Climate and productivity effects on fish mercury (THg) in subarctic were studied.
- THg concentration and bioaccumulation rates of 6 fish species muscle were examined.
- Temperature and productivity were correlated with THg concentration in 4 species.
- Higher climate-productivity was linked to faster THg bioaccumulation in 2 species.
- Joint environmental stressors should be considered in future THg studies on fish.

GRAPHICAL ABSTRACT



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ABSTRACT

Climate change is resulting in increased temperatures and precipitation in subarctic regions of Europe. These changes are extending tree lines to higher altitudes and latitudes, and enhancing tree growth enabling intensification of forestry into previously inhospitable subarctic regions. The combined effects of climate change and land-use intensification extend the warm, open-water season in subarctic lakes and increase lake productivity and may also increase leaching and methylation activity of mercury within the lakes. To assess the joint effects of climate and productivity on total mercury (THg) bioaccumulation in fish, we conducted a space-for-time substitution study in 18 tributary lakes of a subarctic watercourse forming a gradient from cold pristine oligotrophic lakes in the northern headwaters to warmer and increasingly human-altered mesotrophic and eutrophic systems in the southern lower reaches.
Increasing temperature, precipitation, and lake productivity were predicted to elevate length- and age-adjusted THg concentrations, as well as THg bioaccumulation rate (the rate of THg bioaccumulation relative to length or age) in muscle tissue of European whitefish (*Coregonus lavaretus*), vendace (*Coregonus albula*), perch (*Perca fluviatilis*), pike (*Esox lucius*), roach (*Rutilus rutilus*) and ruffe (*Gymnocephalus cernua*).
A significant positive relationship was observed between age-adjusted THg concentration and lake climate-productivity in vendace ($r^2 = 0.50$), perch ($r^2 = 0.51$), pike ($r^2 = 0.55$) and roach ($r^2 = 0.61$). Higher climate-productivity values of the lakes also had a positive linear (pike; $r^2 = 0.40$ and whitefish; $r^2 = 0.72$) or u-shaped (perch; $r^2 = 0.64$ and ruffe; $r^2 = 0.50$) relationship with THg bioaccumulation rate.
Our findings of increased adjusted THg concentrations in planktivores and piscivores reveal adverse effects of warming climate and increasing productivity on these subarctic fishes, whereas less distinct trends in THg bioaccumulation rate suggest more complex underlying processes. Joint environmental stressors such as climate and productivity should be considered in ongoing and future monitoring of mercury concentrations.
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* Corresponding author.
E-mail address: salla.a.ahonen@helsinki.fi (S.A. Ahonen).

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1. Introduction

Recent climate change has greatly impacted Arctic regions, resulting in temperature and precipitation increases above global average (IPCC, 2015). Furthermore, increased global interests in the Arctic and subarctic regions are intensifying the resource use in terrestrial and aquatic ecosystems (Huntington et al., 2007; Wrona et al., 2013; Gordon et al., 2018). Enhanced vegetation growth and tree-line extension in the subarctic, due to climate change, are enabling and intensifying forestry associated with peatland ditching and site-preparation practices that expose the mineral soil and increase nutrient and organic carbon runoff (Serreze et al., 2000; Bates et al., 2008; Nieminen et al., 2015). Nutrient and heavy metal loadings are often connected to the concentrations of dissolved organic carbon (DOC), as DOC acts as a carrier of them from catchments to aquatic ecosystem (Porvari and Verta, 2003; de Wit et al., 2014). In freshwater ecosystems, these environmental changes result in an extended open-water season, increased load and concentrations of nutrients and DOC leading to elevated productivity (Karlsson et al., 2009; Sarkkola et al., 2009; Nieminen et al., 2015).

Mercury (Hg) is a toxic heavy metal, the methylated form (MeHg) of which bioaccumulates in body tissues of fishes (Morel et al., 1998; Sonesten, 2003a; Trudel and Rasmussen, 2006) and biomagnifies to higher trophic levels in food webs (Jernelöv and Lann, 1971; Morel et al., 1998; Lavoie et al., 2013). Hg originates from natural and anthropogenic sources, but the relative proportion of anthropogenically derived Hg has increased since the Industrial Revolution (Pacyna et al., 2010; UNEP, 2013). In relatively pristine subarctic regions, Hg primarily enters aquatic ecosystems through atmospheric deposition of inorganic Hg (Schroeder and Munthe, 1998; AMAP, 2011; Stern et al., 2012).

Methylation of inorganic Hg to MeHg can occur in lake sediment, water column, and in catchment soils, principally by sulphur-oxidizing bacteria (Morel et al., 1998). In subarctic lakes, the production of MeHg predominantly occurs in the anoxic hypolimnion and lake sediment (Eckley and Hintelmann, 2006), whilst demethylation dominates in the oxygenated photic zone (Morel et al., 1998; Chen et al., 2003). The influence of climate change on methylation-demethylation processes and Hg concentrations in the water and aquatic biota is highly complex and likely system dependent. Mercury methylation rate is positively related to temperature (Bodaly et al., 1993), whereas negative or no connection has been found between demethylation and temperature (Bodaly et al., 1993; Canário et al., 2007). Yang et al. (2016) observed higher MeHg production in warm (+8 °C) versus cold (−2 °C) soils, suggesting an enhanced production of MeHg in the warming Arctic soils. A positive relationship between temperature in lake epilimnion and total mercury (THg) levels in fish was also observed in the Canadian subarctic (Evans et al., 2005, 2005), possibly due to temperature-induced higher net methylation rate and Hg inputs from watersheds.

Climate change is resulting in increased leaching of nutrients and DOC in the subarctic, and this may also be expected to increase the availability of Hg in the water column and aquatic food webs, as DOC is a carrier of Hg (Grigal, 2002; Rydberg et al., 2010). Elevated DOC load is also causing browning and lower light penetration in the water column, which expands the anoxic zone suitable for methylation (Lehnherr, 2014; Klapstein et al., 2018). Decomposition of organic matter further stimulates the activity of microbial methylation (Bravo et al., 2017), but DOC also binds Hg, reducing its bioavailability for methylation (Ullrich et al., 2001). The origin of the organic matter has influence. For example, lakes with high levels of algal-derived organic matter exhibit increased bacterial activity and methylation rate. In addition, terrigenous organic matter has been connected to ultimately higher MeHg concentrations (Bravo et al., 2017). Thus, modification of terrestrial soil, due to intensive forestry, can elevate the availability of MeHg to aquatic systems and wildlife (Morel et al., 1998; Lehnherr, 2014).

Studies assessing how lake productivity influences mercury concentrations and bioaccumulation rates of fishes have, to date, proved inconclusive. In a worldwide study, total phosphorus level was found to have a negative connection to MeHg biomagnification, possibly through biomass dilution of THg in primary producers as well as elevated growth rate of organisms at higher trophic levels (Lavoie et al., 2013). A multi-lake study from northeastern North America showed that relatively low productivity in lakes was connected to higher Hg bioaccumulation in food webs (Chen et al., 2005). However, a positive connection between phosphorus level and biomagnification of THg in food webs was found in a Canadian lake (Kidd et al., 2012), and fish communities especially in lakes with forested watersheds were shown to have high concentrations of Hg (Sonesten, 2003b; Chen et al., 2005).

The highest THg concentrations in aquatic biota are generally found in top predators with long lifespans (Cabana and Rasmussen, 1994). Mercury in the water column is integrated into pelagic primary producers by active transport and passive diffusion (Pickhardt and Fisher, 2007), whereas organisms at higher trophic levels predominantly gather Hg from their diet (Hall et al., 1997). The proportion of MeHg of the THg in tissues increases with trophic level, and typically makes up over 90% of the THg in fish muscle (Watras et al., 1998; Madenjian et al., 2016). Several traits including foraging guilds, thermal guilds, and life-history can all influence the THg concentrations in fish muscle (Trudel and Rasmussen, 2006; Karimi et al., 2016; Thomas et al., 2016). Fish species foraging on pelagic prey typically have higher THg concentrations than benthic feeding fishes, due to higher MeHg concentrations often found in zooplankton than littoral benthic prey (Power et al., 2002; Karimi et al., 2016; Thomas et al., 2016). Moreover, increasing growth rate reduces THg concentrations in fish through growth dilution, while starvation has the opposite effect (Simoneau et al., 2005; Keva et al., 2017).

The subarctic offers a unique region to evaluate the joint effects of climatic and land-use variables on THg concentrations and bioaccumulation rate in fish. Subarctic watercourses represent a gradient ranging from pristine cold and oligotrophic lakes to increasingly human-modified and warmer more productive lakes. This gradient from north to south encompasses the distribution limits of several fish species, as fish communities are dominated by salmonids to percids to cyprinids along increasing air temperature (Hayden et al., 2017). Climate related changes in consumer community structure may also have a significant bearing on the Hg dynamics in subarctic lakes. Climate change and associated increase in lake productivity are modifying the fish communities in subarctic lakes as warmer water adapted species are expanding their ranges northwards (Rolls et al., 2017). The presence of novel fish species assemblages modifies consumer community structure, selective foraging, resource competition, and the number of trophic linkages in food webs, all of which can affect Hg burden to piscivores (Thomas et al., 2016). As such, subarctic environmental gradients represent an opportunity to predict future trends of fish communities and create a realistic natural setting to assess climate and productivity effects on THg in fish at a landscape level (Hayden et al., 2017).

We conducted a space-for-time study on a Fennoscandian subarctic watercourse gradient from north to south to determine how THg concentrations and bioaccumulation rates of six fish species are influenced by lake temperature and productivity. The selected fish species are abundant throughout the study area, but differ in their thermal and foraging guilds (Magnuson et al., 1979; Hayden et al., 2013; Thomas et al., 2016). Hg in three generalist species that forage across both pelagic and littoral habitats were assessed: the cold-water adapted European whitefish (*Coregonus lavaretus*) that dominate oligotrophic lakes; the cool-water species Eurasian perch (*Perca fluviatilis*) that dominate mesotrophic lakes, and the warm-water species roach (*Rutilus rutilus*) found in eutrophic lakes (Hayden et al., 2017). Perch undergo ontogenetic dietary shifts from zooplankton to benthic macroinvertebrates, and finally to fish prey, whereas whitefish and roach feed almost exclusively on invertebrate prey (Hayden et al., 2014a). In addition, two specialist taxa were studied: vendace (*Coregonus albula*), a cold-water pelagic zooplanktivore, and ruffe (*Gymnocephalus cernua*), a cool-water specialist benthivore (Hayden et al., 2013; Thomas et al., 2016). Pike (*Esox*

lucius), a cool-water obligate piscivore, were also sampled. This species is primarily associated with littoral habitat and feeds on a wide range of prey fish species (Kahilainen and Lehtonen, 2003). All study species forage visually, but percids and cyprinids are more adapted to feeding under increased turbidity than cold-water salmonids (Rolls et al., 2017). Ruffe, perch, and roach are expanding their ranges towards higher latitudes and altitudes in this area (Hayden et al., 2013, 2014a).

In the current study, we assessed how climate and lake productivity, lake morphometry and catchment properties influence length- and age-adjusted THg concentrations, and the bioaccumulation rate of THg in whitefish, vendace, perch, pike, roach, and ruffe. Using a space-for-time substitution approach, we tested whether THg concentrations in fish were related to lake climate-productivity along a subarctic watercourse with climatic and land-use gradients from north to south (Hypothesis 1). We hypothesized that THg bioaccumulation rates relative to fish length and age (the slope of regression between THg and length or age) would increase along the gradient from north to south (Hypothesis 2), since increase in temperature and productivity of lakes cause shifts in community structure, elevating the amount of pelagic derived energy (Hayden et al., 2017; Leppänen et al., 2017).

2. Material and methods

2.1. The study sites

Eighteen tributary headwater lakes located in the subarctic Tornio-Muonionjoki watercourse were sampled in northern Fennoscandia (Fig. 1). The lakes are distributed along a climate and productivity gradient from northern to southern Lapland where the open water season air temperature, precipitation, and total phosphorus increased by 3.2 °C (ranging from 8.4 to 11.6 °C), 30% (ranging from 197 to 257 mm), and total 45 μg/L (ranging from 3 to 48 μg/L), respectively (Table 1). Conservative climate change models for this region predict increases in temperature and productivity matching the temperature, precipitation, and productivity measured ranges here by the end of this century (IPCC, 2015). Land-use related variables, such as total length of artificial ditches and tree volume in the catchment increased along the same gradient, indicating intensification of land use.

Lake characteristics ranged from clear oligotrophic lakes (<15 μg/L total phosphorus, lakes 1–7, 9) in the north to increasingly mesotrophic (15–29 μg/L total phosphorus, lakes 8, 10–13) and eutrophic lakes (>29 μg/L total phosphorus, lakes 14–18) towards the south (Fig. 1, Table 1). The highest latitude lakes (1–7, 9) are surrounded by mountain birch forest, with low human population density and land use consisting mainly of reindeer herding. Theoretical compensation depth, the depth at which 1% of surface light intensity was detectable, decreases towards the south. The mesotrophic and slightly more colored lake (8) is located below the northern distribution limit of Scots pine (*Pinus sylvestris*) with increasing human population density. The lower latitude mesotrophic lakes (10–13) are located below the northern distribution limit of Norway spruce (*Picea abies*) with higher human population density and forestry as well as nature tourism. The eutrophic lakes (14–18) are characterized by the highest human population density in the catchment among the study lakes, as well as intensive forestry, with associated site-preparation and ditching practices, as the principal forms of land use, enhancing nutrient loading to the lakes, and thus increasing the water color (Jussila et al., 2014).

2.2. Environmental variables

The climate and lake environmental variables were primarily obtained from long-term archives. Geographic location and altitude of each lake were obtained from the National Land Survey of Finland. Average air temperature (°C) and precipitation (mm) during the open water season (June–September) were obtained for each lake from long-term data (1981–2010; Finnish Meteorological Institute, Klein Tank et al., 2002) from six weather stations located through the study area (Fig. 1, Table 1). Total phosphorus and nitrogen data were obtained during the fish sampling or regional monitoring programs of the same year (Lapland Centre for Economic Development, Transport and Environment). The lake morphometric variables included lake area (km^2), maximum depth (m), mean depth (m) and lake volume (m^3) that were obtained from Hertta-database (Finnish Environment Institute) or derived from data of bathymetric maps created by using an echosounder-chartplotter device (LCX-112C GPS-chartplotter: Lowrance, Tulsa, Oklahoma, U.S.A.). Visible light intensity (400–700 nm) was measured in the water column using a LI-COR LI-250A light meter (LI-COR Biosciences, Lincoln, NE, U.S.A.). This data was used to calculate the compensation depth that defined the habitat border between the littoral and pelagic zones. The catchment variables were entirely derived from data archives, where the land-use variables were derived from CORINE Land-Cover data implemented in VALUE-tool (Finnish Environmental Institute). These data were used to calculate catchment area (km^2) and relative proportions (%) of different land-use practices (urban, agriculture, forest, sparse vegetation, wetland and water) in catchment and transformed to area coverages e.g. forest (ha). Tree volume data for the forested area was obtained from the open access data bank (The Natural Resources Institute Finland).

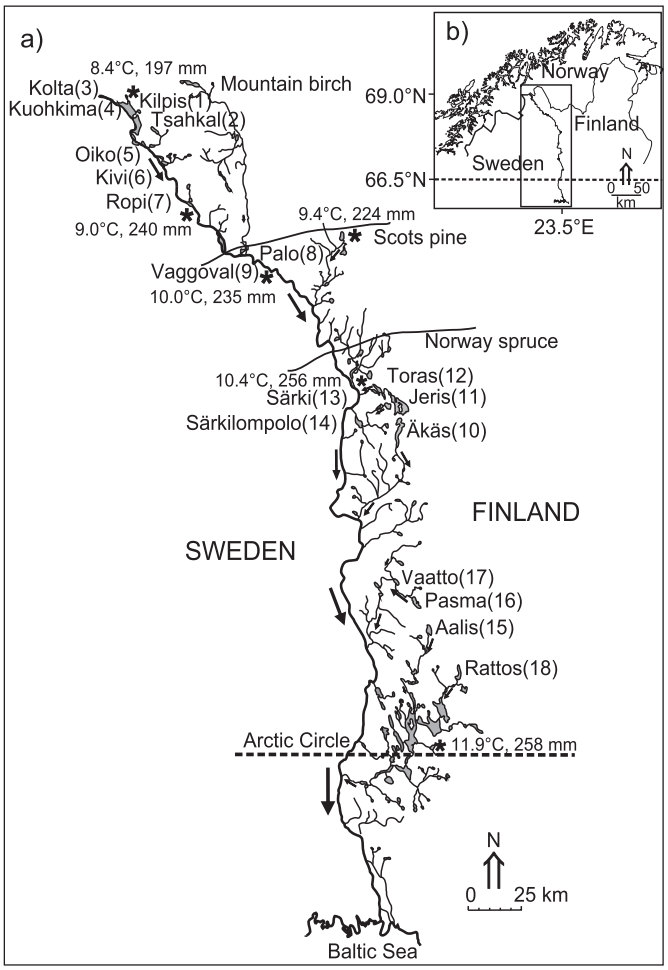

Fig. 1. The location of Tornio-Muoniojoki watercourse (mainstem forms a border between Finland and Sweden) and tributary lakes 1–18 (a) in northern Fennoscandia (b). Lakes are numbered according their PC1 scores (Table 1). Asterisks indicate locations of six weather stations and their open-water season (June–September 1981–2010) mean air temperature (°C) and mean precipitation (mm/season). Arrows are flowing directions of rivers towards south and watercourse finally discharges to the northernmost part of Baltic Sea. Solid lines indicate distribution limit of coniferous trees, where Norway spruce border indicate the start of forestry practices. Mountain birch distribution covers the northernmost lake catchments.

Table 1
Study lakes and selected environmental variables used in principal component analyses (PCA). Altitude (Alt), mean precipitation (Precip) and mean air temperature (Air temp) in open-water season (June–September 1981–2010), total phosphorus (TP), compensation (Comp) and mean (Mean) depths, lake volume (LV), lake area (LA), catchment area (CA), forest (For) and wetland (Wet) percentages in the catchment area, ditching length (km) per 1 km^2 of catchment area (Ditch), tree volume (TV) per hectare of forested and wetland area, catchment-lake area ratio (CA:LA). PCA scores for each lake representing climate and productivity (PC1), lake morphometry (PC2), and catchment magnitude (PC3). Please note that lakes (column No) are ordered by PC1 scores.

No	Lake	Alt (m a.s.l)	Precip (mm)	Air temp (°C)	TP (μg/L)	Depth (m) Comp	Depth (m) Mean	LV (milj m^3)	LA (km^2)	CA (km^2)	For (%)	Wet (%)	Ditch (km/km^2)	TV (m^3/ha)	CA:LA	PC1 (52%)	PC2 (19%)	PC3 (13%)
1	Kilpis	473	197	8.4	4	10	19.4	728.0	37.3	260.35	8.90	4.09	0	18.66	6.98	4.91	−5.62	−0.54
2	Tsahkal	559	197	8.4	5	7.5	9	10.1	1.1	28.76	2.00	5.28	0	7.01	26.15	3.77	0.63	0.16
3	Kolta	490	197	8.4	3	9	1.1	1.4	1.3	31.04	6.90	4.00	0	13.04	23.88	3.33	1.33	0.22
4	Kuohkima	489	197	8.4	3	8	2.6	0.8	0.3	36.17	7.10	2.60	0	12.39	120.58	3.18	1.35	−0.84
5	Oiko	448	218	8.7	7	5	3.1	3.7	1.2	21.24	19.90	10.15	0	20.15	17.70	2.02	1.20	0.46
6	Kivi	445	221	8.7	7	5	2.8	9.7	3.5	165.03	4.70	15.88	0	24.23	47.15	1.99	1.11	−0.60
7	Ropi	399	240	9	10	5	6.9	8.9	1.3	86.80	12.89	18.18	0	23.96	66.77	1.52	0.84	−0.40
8	Palo	346	224	9.4	21	3	0.8	3.0	3.5	134.44	31.50	32.24	0.01	23.29	38.41	0.38	1.56	−0.37
9	Vaggoval	331	236	9.8	12	4	0.5	0.8	1.8	6.03	36.03	38.99	0	32.58	3.35	0.26	1.89	0.53
10	Äkäs	263	256	10.5	18	4.5	3	39.1	13.1	74.04	56.13	12.66	0.15	81.78	5.65	−1.08	−0.54	1.01
11	Jeris	258	256	10.5	25	3.5	3.4	80.8	23.5	98.84	56.02	8.16	0.20	76.11	4.21	−1.19	−1.48	1.07
12	Toras	247	256	10.4	18	3.5	3	14.7	4.9	215.34	62.30	9.90	0.14	80.55	43.95	−1.47	−0.17	−0.05
13	Särki	261	256	10.5	25	1.5	5.1	26.0	5	14.71	58.21	4.68	0.49	74.77	2.94	−1.51	−0.30	1.37
14	Särkilompolo	255	256	10.5	36	2.5	3.7	4.7	1.3	36.93	68.96	6.57	0.23	74.20	28.41	−1.91	0.06	1.05
15	Aalis	177	257	11.4	45	1.5	3.2	19.3	6	72.25	62.25	12.55	6.67	71.09	12.04	−3.35	−0.50	0.58
16	Pasma	164	257	11.3	48	1.5	1.6	13.0	8.4	134.63	65.03	10.59	5.70	71.45	16.03	−3.45	−0.57	0.41
17	Vaatto	156	257	11.2	30	1.5	1.8	4.2	2.3	637.97	65.03	17.28	5.62	70.45	277.38	−3.62	−0.24	−4.96
18	Rattos	118	257	11.6	47	1.5	2.1	8.6	4.1	58.06	72.96	4.27	5.65	78.60	14.16	−3.77	−0.54	0.89

The total length of artificial ditches (km) in the catchment of each lake was analyzed from open map data (National Land Survey of Finland). Tree volume (m^3/ha) and ditch length (km/km^2) variables were calculated for forested and wetland catchments. Analyses were conducted using ESRI ArcMap 10.3.1 software.

2.3. Fish species and field sampling

We sampled six fish species across the 18 study lakes: whitefish, vendace, perch, pike, roach, and ruffe. Whitefish and pike inhabited almost all the study lakes, whereas perch, roach, and ruffe were absent from the northernmost lakes (Table 2). Vendace and roach were abundant in the lower latitudes. Fish were caught over five consecutive days between late-August and September in 2009–2013, to control for potential seasonal THg variation in muscle tissue (Kahilainen et al., 2016; Keva et al., 2017). The target sample size was 30 individuals for each species in each lake, if possible. The fish were caught by gillnet series with eight nets (size: 30 × 1.8 m, knot-to-knot mesh size: 12, 15, 20, 25, 30, 35, 45 and 60 mm), as well as one Nordic multi-mesh gillnet (size 30 × 1.5 m, mesh sizes 5–55 mm) with 12 equidistant panels of 2.5 m. Pike samples were supplemented with hook-and-line sampling due to their comparatively low density. Nets were set overnight (10–12 h) in littoral, profundal, and pelagic habitats in each lake, except shallow full littoral lakes that lack pelagic and profundal zones (Table S1). All these principal habitats were sampled for at least three nights. Fish were removed from nets, euthanized by cerebral concussion causing an immediate death and placed on ice during transport to the laboratory.

All fish were identified to species and total length (±1 mm) and age were determined. For whitefish and vendace, the first left gill arch was dissected and the number of gill rakers counted for morph and species identification purposes (Kahilainen et al., 2011a, 2011b). In this region, whitefish may occur as sympatric morphs having different number of gill rakers, diet, growth, and muscle THg concentrations (Kahilainen and Østbye, 2006; Harrod et al., 2010; Kahilainen et al., 2017). In the study lakes, whitefish occur as a single generalist morph, except Lake Ropi where we selected only the littoral morph for this study (Hayden et al., 2013). Fish ages were determined using the most reliable hard structures for each species including burned cross-sections of otoliths, bones, and scales pressed on polycarbonate plates (Thomas et al., 2016). Two hard structures were simultaneously used to improve the aging accuracy (Kahilainen et al., 2017). Aging of pike was done using cleithrum bone and scale impressions. For perch operculum bone and burned cross-sections of otoliths were used, while for the other species (whitefish, vendace, roach, ruffe) clear and burned cross-sections of otoliths as well as scale impressions were used (Kahilainen et al., 2003, 2011a; Hayden et al., 2014a, 2015). Dorsal muscle tissue samples were taken from each sample, frozen at −20 °C, freeze-dried for 48 h, and grinded with glass rod to fine powder for THg analyses.

2.4. Total mercury analyses

Total mercury (THg) concentrations (μg g^{-1} dry mass) were obtained using a Direct Mercury Analyser (Milestone DMA 80). Two replicates of each sample were analyzed to assess sample variability. A blank control and certified reference material (DORM-4) with known THg concentration (mean ± SD, 0.410 ± 0.055 μg g^{-1}) were included at the beginning and end of each analytical run (mean ± SD, 0.404 ± 0.019 μg g^{-1}, mean recovery 98.6%, n = 390). The THg concentration of each sample used in statistical analyses was derived by calculating the mean value of duplicates after subtracting the mean blank control values (mean ± SD, 0.002 ± 0.001 μg g^{-1}, n = 390). The mean of the duplicates for each sample was accepted for subsequent analyses only when the relative standard deviation (RSD) was <10%. A randomized subset of 30 individuals was chosen from populations where sample size was >30 to make systems comparable.

Due to a strong positive relationship between THg concentration and fish length/age, THg concentrations of fish were corrected for length/age prior to conducting inter-population comparisons (Sonesten, 2003a; Braaten et al., 2017). We first developed linear regression equations for fish THg and length/age of each species in each lake (Table S4–5, Fig. S1–12). The equations were solved for the mean length and age of each species, resulting in a single mean length- and age-corrected THg value for each species within each lake (the average length and age of each species calculated from all studied lakes; Table 2) (Braaten et al., 2017). In populations where the relationship between THg and length or age was below statistically significant level, the length- and age-adjusted values were obtained by calculating the mean of the species-specific THg concentrations near mean length (± 2.5 cm) and age (±1.5 years). Length- and age-adjusted THg concentrations were used for all subsequent statistical analyses examining population level differences.

Table 2
Sample sizes (n) and mean, length- and age-adjusted THg concentrations (μg g^{-1} dry weight) for whitefish (mean length: 23.4 cm, mean age: 4.7 years, n = 467), vendace (mean length: 13.6 cm, mean age: 3.2 years, n = 283), perch (mean length: 16.4 cm, mean age: 4.6 years, n = 361), pike (mean length: 50.4 cm, mean age: 6.2 years, n = 312), roach (mean length: 14.1 cm, mean age: 6.2 years, n = 193) and ruffe (mean length: 9.6 cm, mean age: 4.1 years, n = 409) for each lake. Absence of fish species in the lake is marked as A. Three highest species-specific THg-concentrations for each lake are bolded. Please note that lakes (column No) are ordered by PC1 scores.

No	Lake	Whitefish THg				Vendace THg				Perch THg				Pike THg				Roach THg				Ruffe THg			
		n	mean	length	age	n	mean	length	age	n	mean	length	age	n	mean	length	age	n	mean	length	age	n	mean	length	Age
1	Kilpis	30	0.16	0.19	0.20	A	–	–	–	A	–	–	–	30	0.65	0.49	0.59	A	–	–	–	A	–	–	–
2	Tsahkal	30	**0.43**	0.39	**0.48**	A	–	–	–	A	–	–	–	A	–	–	–	A	–	–	–	A	–	–	–
3	Kolta	30	0.18	0.17	0.18	A	–	–	–	A	–	–	–	16	0.59	0.72	0.64	A	–	–	–	A	–	–	–
4	Kuohkima	30	0.36	0.34	0.31	A	–	–	–	A	–	–	–	18	0.99	–	–	A	–	–	–	A	–	–	–
5	Oiko	30	**0.47**	**0.46**	**0.47**	A	–	–	–	A	–	–	–	27	1.68	**1.66**	**1.56**	1	–	–	–	30	0.40	0.34	0.35
6	Kivi	30	0.40	**0.45**	0.39	A	–	–	–	30	**0.80**	0.36	0.31	18	1.64	1.37	1.22	A	–	–	–	30	**0.60**	**0.57**	**0.44**
7	Ropi	30	0.22	0.23	0.21	A	–	–	–	2	0.30	–	–	22	0.51	0.76	0.85	1	–	–	–	21	0.51	0.53	**0.44**
8	Palo	30	0.23	0.17	0.22	30	0.24	0.19	0.29	30	0.27	0.26	0.29	15	0.60	0.63	0.76	5	0.30	–	–	30	0.21	0.21	0.27
9	Vaggoval	30	0.24	0.26	0.37	A	–	–	–	30	0.33	0.34	0.44	27	0.72	0.90	0.89	1	–	–	–	28	**0.52**	0.52	0.52
10	Äkäs	30	0.17	0.34	0.17	30	0.28	0.29	0.29	30	0.29	0.28	0.36	6	0.78	0.75	1.00	30	0.21	0.23	0.21	30	0.17	0.12	0.17
11	Jeris	30	0.20	0.20	0.27	30	0.26	0.27	0.24	30	0.25	0.31	0.43	4	0.47	–	–	A	–	–	–	30	0.22	0.21	0.25
12	Toras	30	0.23	0.21	0.25	30	0.28	0.30	0.32	30	0.31	0.35	0.38	3	0.82	–	–	30	**0.42**	0.40	0.41	30	0.35	0.32	0.36
13	Särki	30	0.21	0.19	0.24	30	0.18	0.17	0.17	30	0.22	0.24	0.20	1	–	–	–	A	–	–	–	30	0.15	0.11	0.16
14	Särkilompolo	30	0.16	0.14	0.15	30	0.54	**0.59**	0.40	30	0.30	0.33	0.31	17	1.20	1.24	1.31	5	0.33	–	0.32	30	0.07	0.08	0.11
15	Aalis	A	–	–	–	30	**0.46**	**0.57**	**0.45**	29	0.75	**0.78**	**0.76**	30	**1.81**	1.09	**1.62**	30	0.37	0.37	0.38	30	**0.55**	**0.60**	**0.56**
16	Pasma	30	0.27	0.26	0.35	30	0.35	0.30	0.36	30	0.59	0.61	0.60	24	**2.17**	**1.81**	1.44	30	**0.42**	**0.49**	**0.44**	30	0.27	0.36	0.32
17	Vaatto	14	**0.45**	**0.46**	**0.48**	13	**0.49**	0.21	**0.64**	30	**1.03**	**1.07**	**0.91**	28	**2.36**	**2.36**	**1.81**	30	**0.56**	**0.58**	**0.54**	30	0.48	0.55	0.43
18	Rattos	3	0.26	–	–	30	**0.47**	**0.68**	**0.57**	30	**1.01**	**1.08**	**0.92**	26	1.64	1.57	**1.56**	30	**0.60**	**0.59**	**0.53**	30	0.39	**0.57**	0.40

2.5. Statistical analyses

Collinearity among environmental variables was first tested with linear regression analyses, and variables with $r^2 < 0.70$ were chosen for the further analyses. Principal component analysis (PCA) was used for the remaining variables to receive PC score for each study lake along environmental gradients using PC components. PC components explaining >10% of the variation were considered relevant for further evaluation of trends with THg concentrations and bioaccumulation rates. Lake PC scores were negative transformed (*−1) for THg analyses, because their connections to warmer climate and increased lake productivity were visually clearer compared to original scores (for original PC scores, see Fig. 2, Table 1).

Statistical testing of Hypothesis 1: Linear regression analysis was used to test the relationship between length- or age-adjusted THg concentration and lake PC scores. Lake-specific populations of each species were used for regression analyses only if the sample size was greater or equal to six.

Statistical testing of Hypothesis 2: The relationship between THg bioaccumulation rate (i.e. the slopes of significant linear regressions between THg, and length or age) and lake PC scores representing different environmental gradients were also tested using simple linear regression analysis. In addition, non-linear quadratic models were tested for species having visually non-linear relationship between PC scores and THg bioaccumulation rate. Only populations with significant bioaccumulation of THg relative to length or age could be chosen for the subsequent analyses of the bioaccumulation rate.

The statistical significance limit to reject the null hypothesis (alpha) was 0.05 for all analyses. Adjusted R-squared (adj.r^2) values were used for interpreting the significant results:

$$r^2_{adj.} = 1 - \left[\frac{(1-r^2)(n-1)}{n-k-1}\right]$$

where n is the sample size, and k is the number of explanatory variables in the model, excluding the constant term. For simplicity, term r^2 is used in tables. All analyses were conducted using SPSS Statistics 24 (IBM Corp., Armonk, NY, U.S.A.) or R (R Core Team, 2017).

3. Results

3.1. Environment, lake morphometry and catchment variation

The collinearity of environmental variables was considerable due to their nature, and thus we used the remaining 14 variables in PCA (Table 1; see the excluded variables in Table S1). The first three PC components explained a total of 84% of the variation among lakes (Fig. 2). PC1 explained 52% of the variation and included variables related to climate and lake productivity (i.e. altitude, air temperature, precipitation, total phosphorus, and compensation depth) and to catchment related variables (i.e. forest percentage, ditch length and tree volume in the catchment area; see loadings in the supplementary Table S2). PC1 lake scores were correlating negatively to lake climate-productivity (Fig. 2, Table 1). The second axis (PC2) explained 19% of the variance, represented mainly lake morphometric characteristics (i.e. mean depth, lake volume, lake and catchment areas, wetland proportion in catchment, see Table S2) and was negatively correlated with lake size (Fig. 2, Table 1). The third axis (PC3) explained 13% of the variance in lakes including catchment properties (e.g. catchment-lake ratio and catchment area, as well as ditch length, wetland, and tree proportions in catchment with much lower importance, see Table S2) with values correlating negatively to catchment magnitude (Fig. 2, Table 1). Please note, that negative transformed values (*−1) were used for lake scores for the subsequent analyses of the relationship between THg concentration or THg bioaccumulation rate, and PC factors.

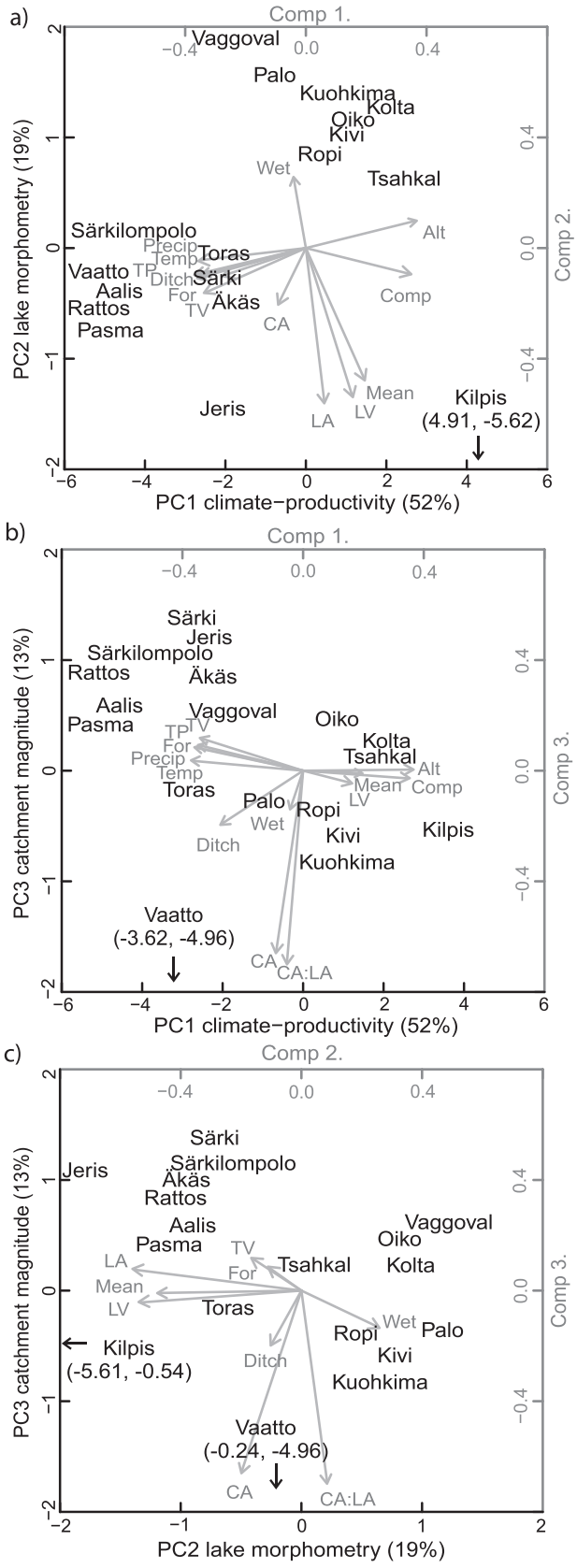

Fig. 2. Results of principal component analysis (PCA) representing the first three axes explaining most of the variation (total 84%) with a total of 14 environmental variables (Table 1) presented in three bi-plots (a, b, c). The first axis (PC1) describes climate-productivity, the second axis (PC2) lake morphometry, and the third axis (PC3) catchment magnitude. The arrow lengths indicate the importance of an environmental variable. For lake scores, see Table 1 and for variable loadings, see Table S2.

3.2. Hypothesis 1: Connection between THg concentrations and lake PC scores

Fish sample sizes were highly variable within species and lakes, partially due to differing distribution limits of each species (Table 2). Whitefish and pike inhabited almost all the study lakes, while other species were absent in the northernmost lakes. THg concentrations varied substantially within species, the highest length- and age-adjusted THg concentrations being found in pike, followed by perch and roach, and the lowest in vendace, ruffe, and whitefish (Table 2). Bioaccumulation of THg relative to length and age varied substantially among fish species (Tables S4–5, Figs. S1–12). THg bioaccumulation relative to length and age were significant for vendace, perch, pike, and ruffe in all or nearly all of the lakes, whereas whitefish and roach had significant regression coefficients in approximately half of the cases (Table S4–5 and Fig. S1–12). Significant coefficients were positive for all the species, except for whitefish having a significant negative relationship between THg concentration and length in two northernmost lakes (Fig. S1).

Lake-specific length- and age-adjusted THg concentrations displayed statistically significant relationships with PC1 (climate-productivity), but were unrelated to either PC2 (lake morphometry) or PC3 (catchment magnitude; see Table 3 and Table S3, Fig. 3 and Figs. S13–14). Length-adjusted THg had a significant positive relationship with lake PC1 score for perch (adj.$r^2 = 0.50$, $p < 0.01$), pike (adj.$r^2 = 0.29$, $p < 0.05$) and roach (adj.$r^2 = 0.57$, $p = 0.05$), whereas relations were below statistically significant levels for whitefish, ruffe and vendace. Significant positive relationships between age-adjusted THg concentration and PC1 were found for vendace (adj.$r^2 = 0.50$, $p < 0.05$), perch (adj.$r^2 = 0.51$, $p < 0.01$), pike (adj.$r^2 = 0.55$, $p < 0.005$) and roach (adj.$r^2 = 0.61$, $p < 0.05$), while linear models for whitefish and ruffe were non-significant (Table 3).

3.3. Hypothesis 2: Bioaccumulation rate of THg relative to lake PC scores

Bioaccumulation rate of THg relative to length or age (the slope of linear regression for each lake and species, see Tables S4–5) showed significant relationships with lake PC1 and PC3 scores, but not with PC2 scores (Table 3, Fig. 3, Table S3, Figs. S13–14). Due to low number of populations having statistically significant bioaccumulation of THg relative to length ($n = 3$) or age ($n = 4$), roach was omitted from these analyses.

The bioaccumulation rate of whitefish THg relative to length displayed a positive linear relationship with PC1 (adj.$r^2 = 0.34$, $p < 0.05$) (Table 3, Fig. 3). Non-linear u-shaped relationships were found for perch (adj.$r^2 = 0.73$; $p < 0.005$) and ruffe (adj.$r^2 = 0.50$, $p < 0.05$). In addition, positive linear relationships for vendace and pike were near the significance limit (vendace adj.$r^2 = 0.28$, $p = 0.067$; pike adj. $r^2 = 0.22$ $p = 0.052$). THg bioaccumulation rate relative to age displayed a positive linear relationship with PC1 for whitefish (adj.$r^2 = 0.72$, $p < 0.005$) and pike (adj.$r^2 = 0.40$, $p < 0.01$). A non-linear relationship between THg bioaccumulation rate relative to age and PC1 was evident for perch (adj.$r^2 = 0.64$, $p < 0.005$) and ruffe (adj.$r^2 = 0.50$, $p < 0.05$) (Table 3, Fig. 3). For vendace, the linear relationship was close to being considered significant at the $p = 0.05$ level (adj.$r^2 = 0.41$, $p = 0.053$).

Bioaccumulation rate of THg relative to length for vendace (adj.$r^2 = 0.50$, $p < 0.05$) and perch (adj.$r^2 = 0.55$, $p < 0.01$), and relative to age for pike (adj.$r^2 = 0.23$, $p < 0.05$) (Table 3, Fig. S14) were positively related to lake PC3 score. Moreover, the relationship of PC3 with the bioaccumulation rate relative to age was near the significance limit for vendace (adj.$r^2 = 0.32$, $p = 0.085$) and perch (adj.$r^2 = 0.23$, $p = 0.066$), as well as relative to length for pike (adj.$r^2 = 0.22$, $p = 0.054$).

Table 3
Regression equations, adjusted R-squared (r^2), p-values and F-values with degrees of freedom for each species between lake-specific length−/age-adjusted THg concentration (μg g^{-1} dry weight), as well as accumulation rate (Slope) relative to length and age and PC1 and PC3 scores. Statistically significant ($p < 0.05$) models are bolded and marked with *. Roach was excluded from the analyses of THg accumulation rate due to low sample size. Regressions for PC2 are found as supplementary material in Table S3.

Model	Whitefish	Vendace	Perch	Pike	Roach	Ruffe
THg (length)	$y = -0.004x + 0.276$; $r^2 = -0.06$;	$y = 0.067x + 0.217$; $r^2 = 0.16$;	***y = 0.126x + 0.304; $r^2 = 0.50$;**	***y = 0.108x + 1.179; $r^2 = 0.29$;**	$y = 0.093x + 0.183$; $r^2 = 0.57$;	$y = 0.006x + 0.352$; $r^2 = -0.08$;
~ PC1	$p = 0.738$; $F_{1,14} = 0.12$	$p = 0.135$; $F_{1,8} = 2.76$	**$p = 0.006$; $F_{1,10} = 12.01$**	**$p = 0.026$; $F_{1,12} = 6.47$**	$p = 0.050$; $F_{1,4} = 7.68$	$p = 0.831$; $F_{1,12} = 0.05$
THg (age)	$y = 0.000x + 0.296$; $r^2 = -0.07$;	***y = 0.078x + 0.208; $r^2 = 0.50$;**	***y = 0.102x + 0.332; $r^2 = 0.51$;**	***y = 0.104x + 1.159; $r^2 = 0.55$;**	***y = 0.083x + 0.182; $r^2 = 0.61$;**	$y = 0.007x + 0.317$; $r^2 = -0.07$;
~ PC1	$p = 0.972$; $F_{1,14} = 0.00$	**$p = 0.014$; $F_{1,8} = 9.87$**	**$p = 0.005$; $F_{1,10} = 12.53$**	**$p = 0.002$; $F_{1,12} = 16.55$**	**$p = 0.024$; $F_{1,5} = 10.25$**	$p = 0.703$; $F_{1,12} = 0.15$
Slope (length)	***y = 0.002x + 0.007; $r^2 = 0.34$;**	$y = 0.017x + 0.011$; $r^2 = 0.28$;	***y = 0.006x^2 − 0.008x + 0.02;**	$y = 0.005x + 0.037$; $r^2 = 0.22$;	–	***y = 0.005x^2 − 0.006x + 0.02;**
~ PC1	**$p = 0.046$; $F_{1,8} = 5.60$**	$p = 0.067$; $F_{1,8} = 4.50$	**$r^2 = 0.73$; $p = 0.002$; $F_{2,8} = 14.3$**	$p = 0.052$; $F_{1,12} = 4.65$		**$r^2 = 0.50$; $p = 0.018$; $F_{2,9} = 6.55$**
Slope (age)	***y = 0.006x + 0.027; $r^2 = 0.72$;**	$y = 0.024x + 0.029$; $r^2 = 0.41$;	***y = 0.012x^2 − 0.012x + 0.05;**	***y = 0.025x + 0.185; $r^2 = 0.40$;**	–	***y = 0.004x^2 − 0.009x + 0.033;**
~ PC1	**$p = 0.002$; $F_{1,7} = 21.74$**	$p = 0.053$; $F_{1,6} = 5.78$	**$r^2 = 0.64$; $p = 0.004$; $F_{2,9} = 10.7$**	**$p = 0.009$; $F_{1,12} = 9.58$**		**$r^2 = 0.50$; $p = 0.018$; $F_{2,9} = 6.49$**
THg (length)	$y = 0.036x + 0.274$; $r^2 = 0.17$;	$y = -0.032x + 0.354$;	$y = 0.094x + 0.508$; $r^2 = 0.19$;	$y = 0.170x + 1.145$; $r^2 = 0.17$;	$y = 0.030x + 0.433$; $r^2 = 0.05$;	$y = 0.053x + 0.362$; $r^2 = 0.12$;
~ PC3	$p = 0.064$; $F_{1,14} = 4.03$	$r^2 = -0.01$; $p = 0.356$; $F_{1,8} = 0.96$	$p = 0.089$; $F_{1,10} = 3.56$	$p = 0.080$; $F_{1,12} = 3.66$	$p = 0.321$; $F_{1,4} = 1.28$	$p = 0.120$; $F_{1,12} = 2.80$
THg (age)	$y = 0.035x + 0.293$; $r^2 = 0.14$;	$y = 0.049x + 0.376$; $r^2 = 0.32$;	$y = 0.067x + 0.496$; $r^2 = 0.13$;	$y = 0.062x + 1.145$; $r^2 = -0.02$;	$y = 0.030x + 0.399$; $r^2 = 0.17$;	$y = 0.029x + 0.327$; $r^2 = 0.05$;
~ PC3	$p = 0.083$; $F_{1,14} = 3.48$	$p = 0.051$; $F_{1,8} = 5.26$	$p = 0.134$; $F_{1,10} = 2.66$	$p = 0.420$; $F_{1,12} = 0.70$	$p = 0.194$; $F_{1,5} = 2.25$	$p = 0.221$; $F_{1,12} = 1.66$
Slope (length)	$y = 0.000x + 0.005$; $r^2 = -0.12$;	***y = 0.016x + 0.047; $r^2 = 0.50$;**	***y = 0.011x + 0.035; $r^2 = 0.55$;**	$y = 0.009x + 0.036$; $r^2 = 0.22$;	–	$y = 0.004x + 0.042$; $r^2 = -0.02$;
~ PC3	$p = 0.855$; $F_{1,8} = 0.037$	**$p = 0.013$; $F_{1,8} = 10.0$**	**$p = 0.005$; $F_{1,10} = 13.28$**	$p = 0.054$; $F_{1,12} = 4.56$		$p = 0.407$; $F_{1,10} = 0.75$
Slope (age)	$y = 0.003x + 0.026$; $r^2 = -0.06$;	$y = 0.016x + 0.079$; $r^2 = 0.32$;	$y = 0.019x + 0.098$; $r^2 = 0.23$;	***y = 0.038x + 0.177; $r^2 = 0.23$;**	–	$y = 0.000x + 0.046$; $r^2 = -0.10$;
~ PC3	$p = 0.480$; $F_{1,7} = 0.56$	$p = 0.085$; $F_{1,6} = 4.24$	$p = 0.066$; $F_{1,10} = 4.27$	**$p = 0.046$; $F_{1,12} = 4.95$**		$p = 0.905$; $F_{1,10} = 0.02$

4. Discussion

4.1. Main results

Increased THg tissue concentration was found in lakes of warmer climate and increased productivity for most species. Adjusted tissue THg concentrations had significant and steep positive slopes with lake climate-productivity (PC1) for perch and pike. Shallower, but still significant, slopes were observed for vendace and roach, whilst no significant relationships were found for whitefish and ruffe. An increasing THg bioaccumulation rate from north to south was not as robustly supported statistically for warmer and more productive lakes. However, our results showed significant positive slopes between THg bioaccumulation rate and PC1 for whitefish and pike, as well as u-shaped relationships for perch and ruffe. Higher THg bioaccumulation rates were related to increasing catchment magnitude (PC3) for vendace, perch, and pike, whereas no relationship between THg concentration or bioaccumulation rate and lake morphometry (PC2) was found for any species.

4.2. THg concentration links to warming and eutrophication of subarctic lakes

Combined effects of changing climate-productivity and catchment land use in the subarctic study lakes was correlated with significantly elevated THg concentrations of several fish species. Foraging guilds of our study fish species encompassed the full suite of foraging strategies evident in these systems, ranging from top predator pike and mesopredator large perch to pelagic zooplanktivore vendace and small roach and perch, as well as benthic invertivores whitefish and ruffe. The highest increase in THg concentrations (i.e. the slope of the regression between THg and PC1) along the study gradient were found for pike (length: 0.108; age: 0.104) and perch (length: 0.126; age: 0.102), whereas lower increases were found for vendace (age: 0.078) and roach (length: 0.093; age: 0.083). Moreover, ruffe and whitefish were lacking significant trends. Previous studies have shown that higher THg levels in fish are connected to increasing trophic level of fish (e.g. Jernelöv and Lann, 1971; Lavoie et al., 2013), as well as the reliance on pelagic rather than littoral benthic food web (Power et al., 2002; Thomas et al., 2016; Kahilainen et al., 2017). Thus, the foraging habitat and the trophic level of species likely influenced the observed trends in the mercury concentrations.

The eutrophication trend of the study lakes, seen as increasing phosphorus and nitrogen concentration and decreasing compensation depth, is evidently caused by joint effects of increasing temperature and precipitation together with intensifying forestry practices, i.e. clear cutting, site preparation and ditching (Jussila et al., 2014), while the proportion of agriculture in the catchments is very low (<1%). The ultimate source of eutrophication is likely important as sewage and agriculture related eutrophication usually cause dilution of THg in fish muscle, whereas forestry related eutrophication seems to have opposite effect (Sonesten, 2003b). Advancing spring thaw due to temperature rise together with increasing precipitation and human-modified catchments enhance the mobility of organic carbon and direct leaching of DOC and Hg to lakes (Stern et al., 2012). The combination of warming lakes and increasing decomposition of DOC enhance low-oxygen conditions suitable for bacterial methylation of Hg in both terrestrial and lake ecosystems (Evans et al., 2005, 2005; Lehnherr, 2014; Yang et al., 2016). Over six-fold decrease in compensation depth from north to lower latitudes likely relates to decreasing oxygen levels and elevating Hg methylation activity.

The proportion of forested watershed (from 2 to 16%) and artificial ditch kilometers (from 0 to 6.67 km/km^2) in the lake catchment both increase towards lower latitudes with simultaneously increasing THg concentrations of four out of six fish species studied. Despite the increasing forested proportion and ditching length, the increase of tree volume (m^3/ ha) in the catchment stopped below the mesotrophic study

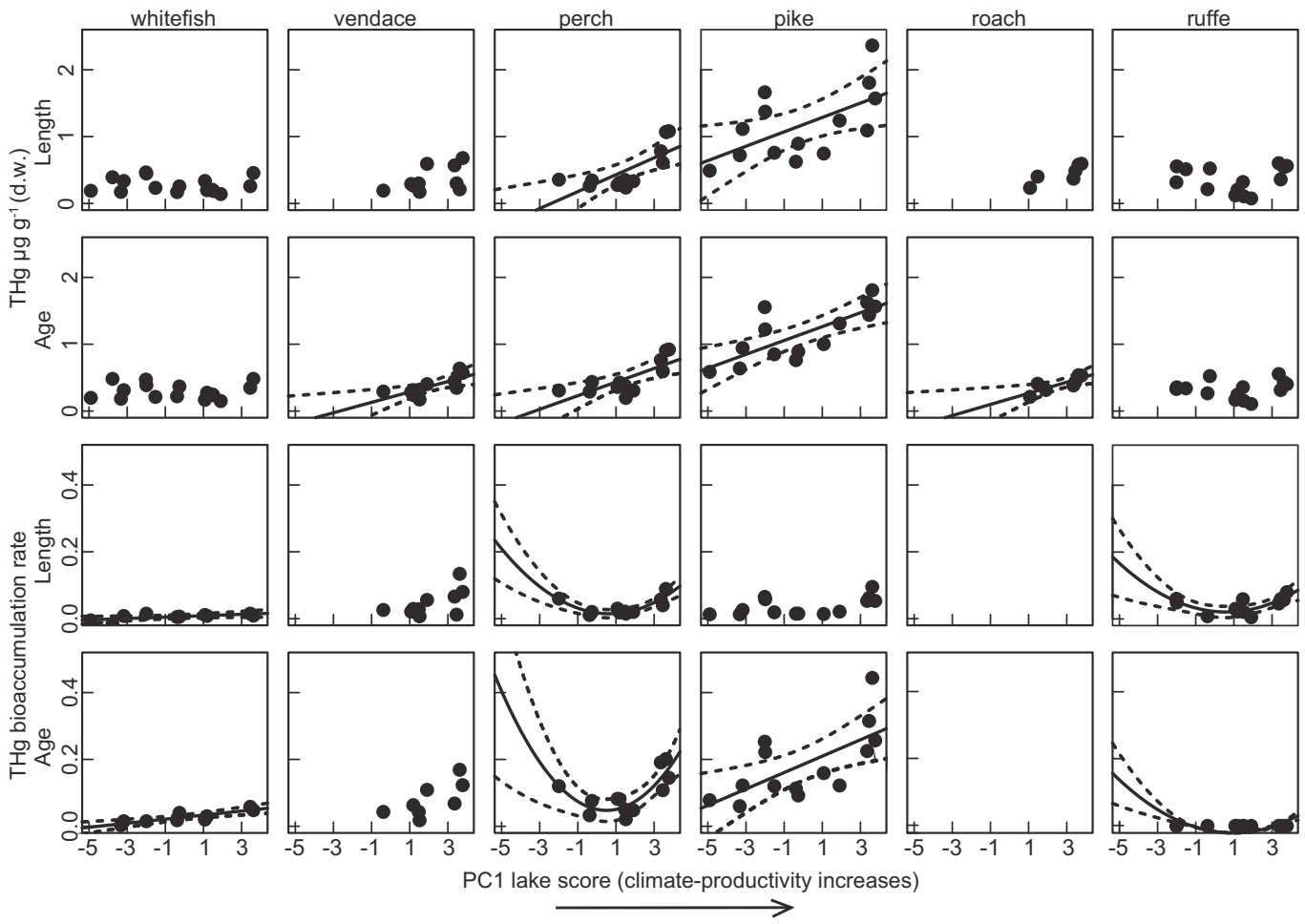

Fig. 3. The relationship between climate-productivity factor (PC1, for clarity opposite lake score values are used) to species-specific length-adjusted THg concentrations (the topmost row) and age-adjusted THg concentrations (the second row), as well as THg bioaccumulation rate relative to length (the third row) and age (the fourth row). Please note that THg concentrations are indicated as dry weight. The regression lines and 95% confidence intervals are added only to statistically significant ($p < 0.05$) cases. For regression equations, see Table 3.

lakes, and has decreased due to extensive clear-cuts in the southern limits of the study region (Jussila et al., 2014). These trends strongly suggest a positive connection between land-use and Hg concentrations in fish. Previous studies have shown that forested peatland catchments and intensified forestry elevate THg concentrations in lake communities and fish (Sonesten, 2003b; Garcia and Carignan, 2005). Forested watersheds and forestry practices increase the load of nutrients, organic matter and Hg to lakes, as well as elevate Hg methylation both in the catchment and lakes (Porvari et al., 2003; Skyllberg et al., 2009; Kronberg et al., 2016, 2016). Such effects of increasing direct leaching of Hg, as well as higher methylation activity due to decomposition of DOC have likely contributed to the increasing fish THg concentrations in our study lakes. The effects of forestry practices on THg in the water and biota are also highly dependent on the forestry operation methods, the cutting season, morphometric characteristics (e.g. slope) of the catchment area, and type of bedrock, complicating the development of robust forestry and water THg concentration relationships at the landscape level (Rask et al., 1998; Eklöf et al., 2014; Kronberg et al., 2016; Ukonmaanaho et al., 2016).

In addition to direct leaching of mercury from clear-cut sites, it is likely that increasing temperature and productivity cause a shift in basal energy sources and have a role in elevated fish THg concentrations. Here, benthic-driven primary production typically dominates in cold oligotrophic lakes, where the pelagic zooplankton have only a single short peak during the mid to late summer (Kahilainen et al., 2003, 2005; Hayden et al., 2014b). In warmer and more productive lakes, the proportion of benthic primary production decreased along as a function of shading and nutrient competition by pelagic phytoplankton and bacterial production (Vadeboncouer et al., 2003; Karlsson et al., 2009). Such effect is evident also in our lakes, where compensation depth decreased from 10 m in the northernmost lakes to 1.5 m in the southernmost lakes with a simultaneous increase in nutrient levels. Previous studies have found much higher THg concentrations in pelagic fish than benthic fish in subarctic lakes (Power et al., 2002; Thomas et al., 2016; Kahilainen et al., 2017). In the subarctic lakes, littoral benthic macroinvertebrates have generally lower mercury content than zooplankton explaining the low THg concentration in benthivorous fish, such as whitefish and ruffe in the study lakes (Kahilainen et al., 2016, 2017; Keva et al., 2017). Zooplanktivores, such as vendace, small roach, and perch, as well as piscivorous fish (pike, large perch) feeding on these pelagic forage fish are often THg enriched (Thomas et al., 2016). However, these foraging guild related THg concentrations are not directly correlated with trends in bioaccumulation rates.

4.3. Links between THg bioaccumulation rate and climate-productivity

THg bioaccumulation rate (the slope of regression between THg and length or age) of whitefish and pike displayed a positive linear relationship with climate-productivity of lakes. However, u-shaped, non-linear relationships were observed for perch and ruffe, with the lowest THg bioaccumulation rate in the mesotrophic lakes. Changes in the THg bioaccumulation rate are less straight-forward than what was seen with

THg concentrations, due to shifts in energy source towards pelagic and fish community structure along the lake gradient (Vadeboncouer et al., 2003; Thomas et al., 2016; Hayden et al., 2017).

For whitefish, THg bioaccumulation relative to length was negative in the northernmost lakes, whereas in more productive lakes bioaccumulation was positive. This change in the direction of the regression resulted in a positive relationship between bioaccumulation rate and lake climate-productivity in the full data. The increasing THg bioaccumulation rate of whitefish southward the climate-productivity gradient is likely related to changes in foraging habitats. In the northernmost oligotrophic lakes, whitefish use pelagic zooplankton mainly in juvenile phase and typically shift to littoral benthic invertebrates in later life stages (Kahilainen et al., 2003). Such ontogenetic diet shifts from THg rich pelagic zooplankton towards THg poor littoral benthic macroinvertebrates evidently reduces the bioaccumulation rate of mercury (Kahilainen et al., 2017). However, whitefish populations in the southern lakes do not demonstrate this ontogenetic shift and continue to feed upon pelagic zooplankton (Heikinheimo et al., 2000), and this ontogenetic change in foraging habitats could be seen in the changing bioaccumulation trends along our study lakes.

THg bioaccumulation rate is increasing for pike in the studied lake gradient from north to south, which is probably related to changes in prey species composition in lower trophic levels, seen as a shift in fish dominance from salmonids to percids and cyprinids towards lower latitudes (Hayden et al., 2017). Pike and other piscivorous fish usually consume the most abundant and easily available prey fish in subarctic lakes (Kahilainen and Lehtonen, 2003). A diet shift of pike and large perch from benthivorous whitefish with relatively low and stable THg concentrations (Hayden et al., 2013; Hayden et al., 2014a; Kahilainen et al., 2017) to other species such as perch, vendace or roach that have increasing THg concentrations from north to south, would likely increase THg bioaccumulation rate of piscivorous pike as well as large perch. Furthermore, larger individuals of aforementioned prey species contained higher THg concentrations that is also important, as prey size of piscivores generally increases with length (Kahilainen and Lehtonen, 2003). Increasing food-chain length towards warmer and eutrophic lakes due to additional pelagic species in lower latitudes (Hayden et al., 2014b; Thomas et al., 2016; Hayden et al., 2017) would also contribute to the higher THg bioaccumulation rate especially in apex predators (pike and large perch).

Vendace, perch, and ruffe did not show evidence of significant linear relationships between THg bioaccumulation rate and lake climate-productivity. However, data revealed u-shaped non-linear relationships for perch and ruffe, with lowest bioaccumulation rate in the mesotrophic lakes. THg concentrations of most fish species increased towards warmer and eutrophic lakes, but the rate of THg bioaccumulation relative to length and age should not be affected, if the growth rate and the THg levels of prey items remained stable. In addition, within the climate and productivity gradients from north to south in the study region, a slight increase in temperature and productivity to a moderate level may increase the growth rate of the study species and lead to growth dilution of THg in fish muscle (Simoneau et al., 2005; Hayden et al., 2014a, 2017). However, further warming and eutrophication beyond the moderate level will obviously lead to reduced growth rate and general increase in THg. This phenomenon is likely responsible for the u-shaped relationships observed for perch and ruffe.

Positive relationships between THg bioaccumulation rate and catchment magnitude (PC3), referring principally to the size of the catchment, were significant for vendace, perch, and pike. However, PC3 axis explained only 13%, of the variation between lakes, considerably less than either PC1 or PC2. Moreover, the PC3 score of Lake Vaatto (lake number 17 in Fig. 1), a large and completely ditched catchment, is clearly different from the other lakes and likely driving significant relationships for the fish species. Since all the other lakes are rather similar with regard to the relative catchment size, caution should be taken when interpreting any possible relationship between PC3 scores and THg bioaccumulation rate, despite the statistically significant regression.

4.4. Human health considerations

High mercury exposure to humans has been linked to damage in neurological, reproductive, immune, and cardiovascular systems (Mergler et al., 2007). The maximum acceptable level set by United Nations (Codex Alimentarius, 1995) of THg in fish muscle tissue for human diet is 0.5 $\mu g\ g^{-1}$ wet mass for non-piscivorous fish (all study species except pike) and 1 $\mu g\ g^{-1}$ for predatory fish (e.g. pike) that correspond to approximately 2.5 and 5 $\mu g\ g^{-1}$ dry mass. Most of the fish species sampled in this current study had THg concentrations below these international health limits, except for a few individual predator perch ($n = 4$) and pike ($n = 4$). The fish with the highest THg concentrations were typically large, old, and located in the most eutrophic and warmest lakes. Thus, local people consuming fishes in the southern lakes, especially pike and perch that are target species of subsistence and recreational fishing, have a higher risk of exposure to THg relative to people consuming fishes caught in northern lakes, where whitefish is the target species (Thomas et al., 2016; Kahilainen et al., 2017; Keva et al., 2017). More attention should also be paid to the fact, that fish consumption in the Arctic and subarctic, e.g. by indigenous Sami people, is generally higher than in lower latitudes and most of the consumption is locally derived fish (Sjölander, 2011). Generally, the increasing THg concentrations of important subsistence fishery species (vendace, perch, and pike) along the climate-productivity gradient of this study from north to south, adds further evidence of adverse effects of eutrophication on the quality of fish as a human diet resource (Taipale et al., 2016).

5. Conclusions

This study revealed a positive linear relationship between adjusted THg concentration in fish and increasing climate-productivity values of subarctic lakes for several fish species. Results indicated that lake eutrophication due to climatic change and intensification of forestry related land use activities have joint effects, elevating THg concentrations in several fish species. Accelerating THg bioaccumulation rate towards warmer and more productive lakes was evident for whitefish and pike, whereas a u-shaped relationship was found for perch and ruffe, indicating that a minor increase in lake temperature and trophic state might lead to growth dilution of Hg for some fish species, whereas loss of clarity and eutrophication lead to elevated THg burden. As evident in the study, THg bioaccumulation and dynamics are both species-specific and lake-specific, and thus the results should be interpreted along the full lake gradient from north to south for each species. Combined effects of climate and land use are highly complex, altering both abiotic and biotic factors in each lake and catchment, and need to be considered in future THg studies of fish. Generally, there is evident need for future studies that examine entire food webs, revealing the pelagic and benthic contributions of THg sources and subsequent biomagnification patterns along climatic and productivity gradients. Such baseline studies are critical for a selection of lakes in the long-term monitoring studies of THg concentrations in the rapidly changing subarctic.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2018.04.436.

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